PERGAMON

Applied Mathematics Letters 15 (2002) 285–291

Applied Mathematics Letters

www.elsevier.com/locate/aml

Modelling of Optically Active Electromagnetic Media

A. MORRO
University of Genoa, DIBE
Via Opera Pia 11a, 16145 Genova, Italy

(Received April 2001; accepted May 2001)

Communicated by G. Lebon

Abstract—By analogy with the DBF model of optically-active electromagnetic media, some generalizations are considered so that memory effects are incorporated. Next, thermodynamic restrictions are derived. It turns out that, via suitable restrictions, the DBF model is compatible with thermodynamics. Also, the analogous terms in a model for nongyrotropic solids are not allowed. The appropriate modification for such a model is indicated.

Keywords—Optical activity, Maxwell's equations, Thermodynamic restrictions.

1. INTRODUCTION

Upon passage through an optical active medium, the direction of polarization of linearly polarized light is rotated. This phenomenon is exhibited by the stereoisomers of organic chemistry and by anisotropic materials such as crystals and ferrites. Indeed, optical rotation was discovered by Arago and Biot in connection with the propagation through anisotropic crystals. Next, Biot found that optical rotation occurs also in organic liquids which are isotropic. The term natural optical activity was then ascribed to such materials thus regarding isotropic optical activity as more natural than optical activity in anisotropic materials. A historical overview of discoveries and modelling about optical activity is given in [1].

In this paper, attention is addressed to constitutive models for natural optical activity. In this regard, we mention that perhaps the most widely applied model is named after Drude-Born-Fedorov (DBF) and is given the form

$$\mathbf{D} = \epsilon\left(\mathbf{E} + \lambda \nabla \times \mathbf{E}\right), \qquad \mathbf{B} = \mu\left(\mathbf{H} + \lambda \nabla \times \mathbf{H}\right), \tag{1}$$

where $\mathbf{D}$ is the electric displacement, $\mathbf{E}$ is the electric field, $\mathbf{B}$ is the magnetic induction, and $\mathbf{H}$ is the magnetic field. These equations are usually considered in the frequency domain, and hence, ϵ, μ, λ are frequency-dependent scalars.

0893-9659/02/$ - see front matter Typeset by $\mathcal{A}\mathcal{M}\mathcal{S}$-TEX
PII: S0893-9659(01)00132-X

The purpose of this paper is to investigate appropriate generalizations of (1), and to cast them in a thermodynamic framework (cf. [2,3]). Formal simplicity indicates that we restrict attention to isotropic media. The generalizations allow for dissipation through memory effects and electric conduction. Also, terms with higher-order derivatives are considered in the standard form for nongyrotropic media. Necessary and sufficient conditions are obtained for compatibility of the models with the statement of second law for nonlocal materials. In particular, we answer the question whether the simultaneous occurrence of λ in (1), is compatible with or is required by thermodynamics. We find that it is consistent with the second law and, furthermore, it is necessary if ϵ and μ are real valued. Also, we show that a standard form of constitutive equations with second-order derivatives is inadequate to account for optical activity through DBF-like terms.

2. SECOND LAW FOR NONLOCAL MATERIALS

Let $\Omega \subset \mathbb{R}^3$ be the region occupied by the body under consideration. For any position vector $\mathbf{x}$, consider functions of $t \in [0,T]$. Let $\mathbf{J}$ be the electric current density and denote by a superposed dot the derivative with respect to the time t. We say that a cycle, at $\mathbf{x}$ and in $[0,T]$, is a set of functions $\mathbf{E},\mathbf{H},\mathbf{D},\mathbf{B},\mathbf{J}$ such that the values at $t=0$ equal the values at $t=T$, i.e., $\mathbf{E}(\mathbf{x},0)=\mathbf{E}(\mathbf{x},T),\dots,\mathbf{J}(\mathbf{x},0)=\mathbf{J}(\mathbf{x},T)$. The second law, for isothermal systems, is taken as the assertion that there is a vector function $\mathbf{N}$ on $\Omega\times[0,T]$ such that the inequality

$$\int_0^T \left[\dot{\mathbf{H}}\cdot\mathbf{B}+\dot{\mathbf{E}}\cdot\mathbf{D}-\mathbf{E}\cdot\mathbf{J}-\nabla\cdot\mathbf{N}\right] dt \le 0 \tag{2}$$

holds for every cycle in $[0,T]$ (cf. [4–6]).

Let $\mathbf{E}$ and $\mathbf{H}$ depend on time t in forms (3) and (4).

$$\mathbf{E}(\mathbf{x},t)=\mathbf{E}_1(\mathbf{x})\cos\omega t+\mathbf{E}_2(\mathbf{x})\sin\omega t, \tag{3}$$

$$\mathbf{H}(\mathbf{x},t)=\mathbf{H}_1(\mathbf{x})\cos\omega t+\mathbf{H}_2(\mathbf{x})\sin\omega t. \tag{4}$$

For linear constitutive equations, also $\mathbf{D}$, $\mathbf{B}$, and $\mathbf{J}$ take the same form. Hence, we have

$$\begin{aligned}\dot{\mathbf{H}}\cdot\mathbf{B}+\dot{\mathbf{E}}\cdot\mathbf{D}-\mathbf{E}\cdot\mathbf{J}&=\omega\left(\mathbf{E}_2\cdot\mathbf{D}_1+\mathbf{H}_2\cdot\mathbf{B}_1\right)\cos^2\omega t-\omega\left(\mathbf{E}_1\cdot\mathbf{D}_2+\mathbf{H}_1\cdot\mathbf{B}_2\right)\sin^2\omega t\\&\quad+\omega\left(\mathbf{E}_2\cdot\mathbf{D}_2-\mathbf{E}_1\cdot\mathbf{D}_1+\mathbf{H}_2\cdot\mathbf{B}_2-\mathbf{H}_1\cdot\mathbf{B}_1\right)\sin\omega t\cos\omega t\\&\quad-\left[\left(\mathbf{E}_1\cdot\mathbf{J}_1\cos^2\omega t+\mathbf{E}_2\cdot\mathbf{J}_2\sin^2\omega t+\left(\mathbf{E}_1\cdot\mathbf{J}_2+\mathbf{E}_2\cdot\mathbf{J}_1\right)\sin\omega t\cos\omega t\right] .\end{aligned}$$

The duration T may be identified with m times $2\pi/\omega$, m being any positive integer. For formal simplicity, we let $m=1$. The function $\mathbf{N}$ is assumed to take on equal values at $t=0,T$. It may happen that the period for $\mathbf{N}$ is $T/2$.

Integration with respect to $t\in[0,T]$ yields

$$\begin{aligned}\int_0^T \left[\dot{\mathbf{H}}\cdot\mathbf{B}+\dot{\mathbf{E}}\cdot\mathbf{D}-\mathbf{E}\cdot\mathbf{J}-\nabla\cdot\mathbf{N}\right] dt&=\pi\left(\mathbf{E}_2\cdot\mathbf{D}_1+\mathbf{H}_2\cdot\mathbf{B}_1-\mathbf{E}_1\cdot\mathbf{D}_2-\mathbf{H}_1\cdot\mathbf{B}_2\right)\\&\quad-\left(\frac{\pi}{\omega}\right)\left(\mathbf{E}_1\cdot\mathbf{J}_1+\mathbf{E}_2\cdot\mathbf{J}_2\right)-\left(\frac{2\pi}{\omega}\right)\nabla\cdot\overline{\mathbf{N}}(\omega),\end{aligned}$$

where $\overline{\mathbf{N}}$ is the mean value of $\mathbf{N}$ in $[0,T]$. This means that (linear) constitutive equations are compatible with the second law (2) only if

$$\mathbf{E}_2\cdot\mathbf{D}_1+\mathbf{H}_2\cdot\mathbf{B}_1-\mathbf{E}_1\cdot\mathbf{D}_2-\mathbf{H}_1\cdot\mathbf{B}_2-\frac{1}{\omega}(\mathbf{E}_1\cdot\mathbf{J}_1+\mathbf{E}_2\cdot\mathbf{J}_2)-\frac{2}{\omega}\nabla\cdot\overline{\mathbf{N}}(\omega)\le 0. \tag{5}$$

Conversely, let $\mathbf{E}$,$\mathbf{H}$,$\mathbf{D}$,$\mathbf{B}$,$\mathbf{J}$ be periodic with period $2\pi/\omega$. Hence, at any point $\mathbf{x}$, the functions $\mathbf{E}(\mathbf{x},t),\dots,\mathbf{J}(\mathbf{x},t)$ are taken in the form $\hat{\mathbf{E}}(\mathbf{x})f_E(t),\dots,\hat{\mathbf{J}}(\mathbf{x})f_J(t)$. The functions $f_E,\dots,f_J$ are assumed to be periodic and continuous in $[0,T]$ and, moreover, $\dot{f}_E, \dot{f}_H$ are assumed piecewise continuous. This implies that $f_E, \dots, f_J$ can be represented by Fourier series, e.g.,

$$f_E(t) = \frac{1}{2}a_0 + \sum_{k=1}^{\infty} \left[a_k \cos k\omega t + b_k \sin k\omega t\right],$$

where $\omega = 2\pi/T$ and

$$a_k = \frac{2}{T}\int_0^T f_E(t)\cos k\omega t\, dt, \qquad b_k = \frac{2}{T}\int_0^T f_E(t)\sin k\omega t\, dt.$$

Moreover, f_E and f_H can be differentiated term by term to obtain $\dot{f}_E$ and $\dot{f}_H$.

Represent $\mathbf{E}(\mathbf{x},t)$ as

$$\mathbf{E}(\mathbf{x},t) = \sum_{k=0}^{\infty} \mathbf{E}_{k1}\cos k\omega t + \mathbf{E}_{k2}\sin k\omega t$$

and the like for $\mathbf{H}, \dots, \mathbf{J}$ while $\mathbf{E}_{k1} = \hat{\mathbf{E}}(\mathbf{x})a_k$, $\mathbf{E}_{k2} = \hat{\mathbf{E}}(\mathbf{x})b_k$. The dependence of $\mathbf{E}_{k1}$ and $\mathbf{E}_{k2}$ on $\mathbf{x}$ is understood and not written. Since (5) holds for every frequency ω, we can write also

$$\begin{aligned} &k\left(\mathbf{E}_{k2}\cdot\mathbf{D}_{k1} + \mathbf{H}_{k2}\cdot\mathbf{B}_{k1} - \mathbf{E}_{k1}\cdot\mathbf{D}_{k2} - \mathbf{H}_{k1}\cdot\mathbf{B}_{k2}\right) \\ &-\left(\frac{1}{\omega}\right)\left(\mathbf{E}_{k1}\cdot\mathbf{J}_{k1} + \mathbf{E}_{k2}\cdot\mathbf{J}_{k2}\right) - \left(\frac{2}{\omega}\right)\nabla\cdot\overline{\mathbf{N}}_k \le 0, \end{aligned} \tag{6}$$

for every integer k and any chosen ω. The index k for $\mathbf{N}$ is a reminder that $\overline{\mathbf{N}}_k = \overline{\mathbf{N}}(k\omega)$.

Upon substituting the pertinent series, we see that the left-hand side of (2), at any $\mathbf{x}$, becomes

$$\begin{aligned} \omega\int_0^T \sum_{h=0}^{\infty}\sum_{k=1}^{\infty} &\{k\left[\mathbf{D}_{h1}\cos h\omega t + \mathbf{D}_{h2}\sin h\omega t\right]\cdot\left[-\mathbf{E}_{k1}\sin k\omega t + \mathbf{E}_{k2}\cos k\omega t\right] \\ &+ \left[\mathbf{B}_{h1}\cos h\omega t + \mathbf{B}_{h2}\sin h\omega t\right]\cdot\left[-\mathbf{H}_{k1}\sin k\omega t + \mathbf{H}_{k2}\cos k\omega t\right]\}\, dt \\ -\int_0^T \sum_{h=0}^{\infty}\sum_{k=0}^{\infty} &\left[\mathbf{J}_{h1}\cos h\omega t + \mathbf{J}_{h2}\sin h\omega t\right]\cdot\left[\mathbf{E}_{k1}\cos k\omega t + \mathbf{E}_{k2}\sin k\omega t\right]\, dt - \int_0^T \nabla\cdot\mathbf{N}\, dt. \end{aligned}$$

Term by term integration, as $t \in [0,T)$, shows that the only nonzero terms are those with $h = k$. Furthermore, integration gives a common factor $T/2 = \pi/\omega$. Hence, we have

$$\sum_{k=1}^{\infty} k\left(-\mathbf{D}_{k1}\cdot\mathbf{E}_{k2} + \mathbf{D}_{k2}\cdot\mathbf{E}_{k1} - \mathbf{B}_{k1}\cdot\mathbf{H}_{k2} + \mathbf{B}_{k2}\cdot\mathbf{H}_{k1}\right) - \frac{1}{\omega}\sum_{k=1}^{\infty}\left(\mathbf{J}_{k1}\cdot\mathbf{E}_{k1} + \mathbf{J}_{k2}\cdot\mathbf{E}_{k2}\right) - \frac{2}{\omega}\nabla\cdot\overline{\mathbf{N}}_k.$$

In view of (6), we conclude that (2) holds for any pair of functions $\mathbf{E}(t), \mathbf{H}(t)$ which are continuous with piecewise continuous derivatives.

3. GENERALIZATION OF THE DBF MODEL

Given ϵ^0 and $\epsilon' \in L^1(\mathbb{R}^+)$, possibly dependent on the position $\mathbf{x} \in \Omega \subset \mathbb{R}^3$, we denote by $\epsilon*$ a generalized convolution in the form

$$(\hat{\epsilon} * \mathbf{E})(\mathbf{x},t) = \epsilon^0(\mathbf{x})\mathbf{E}(\mathbf{x},t) + \int_0^\infty \epsilon'(\mathbf{x},u)\mathbf{E}(\mathbf{x},t-u)\, du.$$

As a generalization of the DBF model (1), we consider the constitutive equations

$$\begin{aligned} \mathbf{D} &= \hat{\epsilon} * \mathbf{E} + \hat{\alpha} * \nabla\times\mathbf{E}, \\ \mathbf{B} &= \hat{\mu} * \mathbf{H} + \hat{\beta} * \nabla\times\mathbf{H}, \\ \mathbf{J} &= \hat{\sigma} * \mathbf{E} + \hat{\gamma} * \nabla\times\mathbf{E}, \end{aligned}$$

where electric conductivity and (fading) memory effects are allowed. The dependence of $\epsilon^0, \dots, \gamma^0$ and of the functions $\epsilon', \dots, \gamma'$ on the position means that the material is allowed to be inhomogeneous. There is no conceptual difficulty in replacing the scalars $\epsilon^0, \dots, \gamma^0$, $\epsilon'(u), \dots, \gamma'(u)$ with second-order tensors but it is for formal simplicity that we restrict attention to the isotropic case.

Let $\mathbf{E}$ and $\mathbf{H}$ depend on time t in forms (3) and (4). Upon substitution, we have

$$\mathbf{D} = \mathbf{D}_1 \cos\omega t + \mathbf{D}_2 \sin\omega t,$$

where

$$\mathbf{D}_1 = \left(\epsilon^0 + \epsilon'_c\right)\mathbf{E}_1 + \epsilon'_s\mathbf{E}_2 + \left(\alpha^0 + \alpha'_c\right)\nabla\times\mathbf{E}_1 + \alpha'_s\nabla\times\mathbf{E}_2, \tag{7}$$

$$\mathbf{D}_2 = \left(\epsilon^0 + \epsilon'_c\right)\mathbf{E}_2 - \epsilon'_s\mathbf{E}_1 + \left(\alpha^0 + \alpha'_c\right)\nabla\times\mathbf{E}_2 - \alpha'_s\nabla\times\mathbf{E}_1, \tag{8}$$

and the like for $\mathbf{B}$ and $\mathbf{J}$. The subscripts c, s denote the cosine and sine Fourier transforms.

Detailed calculations are more profitably performed by using the phaser representation. Letting

$$\mathcal{E} = \mathbf{E}_1 + i\mathbf{E}_2,$$

we have

$$\mathbf{E}_1 \cos\omega t + \mathbf{E}_2 \sin\omega t = \Re[\mathcal{E}\exp(-i\omega t)],$$

where $\Re$ denotes the real part. Upon substitution for $\mathbf{E}$ in the expression for $\mathbf{D}$, we have

$$\mathbf{D}(t) = \Re[(\epsilon\mathcal{E} + \alpha\nabla\times\mathcal{E})\exp(-i\omega t)],$$

where

$$\epsilon = \epsilon^0 + \int_0^\infty \epsilon'(u)\exp(i\omega u)\,du, \qquad \alpha = \alpha^0 + \int_0^\infty \alpha'(u)\exp(i\omega u)\,du.$$

If we let

$$\mathcal{D} = \mathbf{D}_1 + i\mathbf{D}_2 := \epsilon\mathcal{E} + \alpha\nabla\times\mathcal{E}$$

and observe that

$$\epsilon(\omega) = \epsilon^0 + \epsilon'_c(\omega) + i\epsilon'_s(\omega), \qquad \alpha(\omega) = \alpha^0 + \alpha'_c(\omega) + i\alpha'_s(\omega),$$

representations (7),(8) are recovered. By the same token, we find that

$$\mathcal{B} = \mu\mathcal{H} + \beta\nabla\times\mathcal{H}, \qquad \mathcal{J} = \sigma\mathcal{E} + \gamma\nabla\times\mathcal{E}.$$

Maxwell's equations then take the form

$$\nabla\times\mathcal{E} = i\omega\mathcal{B}, \qquad \nabla\times\mathcal{H} = -i\omega\mathcal{D} + \mathcal{J}. \tag{9}$$

Upon substitution, we have

$$\mathcal{D} = \epsilon\mathcal{E} + i\omega\alpha\mathcal{B}, \qquad \mathcal{B} = \mu\mathcal{H} - i\omega\beta\mathcal{D} + \beta\mathcal{J}, \qquad \mathcal{J} = \sigma\mathcal{E} + i\omega\gamma\mathcal{B}.$$

The solution for $\mathcal{D}, \mathcal{B}, \mathcal{J}$ in terms of $\mathcal{E}$ and $\mathcal{H}$ turns out to be

$$\mathcal{D} = \frac{\epsilon(1 - i\omega\beta\gamma) + i\omega\alpha\beta\sigma}{\delta}\mathcal{E} + \frac{i\omega\alpha\mu}{\delta}\mathcal{H}, \tag{10}$$

$$\mathcal{B} = \frac{\beta\sigma - i\omega\beta\epsilon}{\delta}\mathcal{E} + \frac{\mu}{\delta}\mathcal{H}, \tag{11}$$

$$\mathcal{J} = \frac{\sigma(1 - \omega^2\alpha\beta) + \epsilon(\omega^2\beta\gamma - 1)}{\delta}\mathcal{E} + \frac{i\omega\mu}{\delta}\mathcal{H}, \tag{12}$$

where $\delta = 1 - \omega^2\alpha\beta - i\omega\beta\gamma$. Hence, we obtain $\mathbf{D}_1, \mathbf{D}_2, \dots, \mathbf{J}_2$ in terms of $\mathbf{E}_1, \mathbf{E}_2$, $\mathbf{H}_1, \mathbf{H}_2$.

As with any thermodynamic analysis, we have to ascertain which quantities can be chosen arbitrarily. Equations (9) may be viewed as the definitions of $\nabla\times\mathcal{E}$ and $\nabla\times\mathcal{H}$ in that, for any triplet of vectors $\mathcal{B}, \mathcal{D}, \mathcal{J}$, the values of $\nabla\times\mathcal{E}$ and $\nabla\times\mathcal{H}$ as given by (9) satisfy Maxwell's equations. Further, the remaining equations for $\nabla\cdot\mathcal{B}$ and $\nabla\cdot\mathcal{D}$ are viewed as constraints on the derivatives of $\mathcal{B}$ and $\mathcal{D}$. Accordingly, Maxwell's equations allow the vector values $\mathcal{D}, \mathcal{B}, \mathcal{J}$ to be chosen arbitrarily at any point $\mathbf{x}\in\Omega$. In turn, once we refer to the constitutive equations in form (10)–(12), the vector values $\mathcal{E}$ and $\mathcal{H}$ can be chosen arbitrarily at any point $\mathbf{x}\in\Omega$.

For definiteness, we now examine in detail particular cases.

4. RESTRICTIONS FOR A DISSIPATIVE DBF MODEL

Let $\sigma, \gamma = 0$. Hence, (10) and (11) simplify to

$$\mathcal{D} = a\mathcal{E} + b\mathcal{H}, \qquad \mathcal{B} = c\mathcal{E} + d\mathcal{H}, \tag{13}$$

where

$$a = \frac{\epsilon}{1-\omega^2\alpha\beta}, \qquad b = \frac{i\omega\alpha\mu}{1-\omega^2\alpha\beta}, \qquad c = \frac{-i\omega\beta\epsilon}{1-\omega^2\alpha\beta}, \qquad d = \frac{\mu}{1-\omega^2\alpha\beta}. \tag{14}$$

Really, we look for conditions on generic coefficients $a, b, c, d \in \mathbb{C}$ in (13) and next we particularize to the coefficients (14).

Inequality (5), with $\mathbf{J} = 0$, can be written as

$$\Re\{i\omega\mathcal{E}^* \cdot \mathcal{D} + i\omega\mathcal{H}^* \cdot \mathcal{B}\} \le 0,$$

where the superscript $*$ means complex conjugate. Hence, because of (13), we have

$$\begin{aligned} -a_I\left(\mathbf{E}_1^2 + \mathbf{E}_2^2\right) - d_I\left(\mathbf{H}_1^2 + \mathbf{H}_2^2\right) - (b_I + c_I)\left(\mathbf{E}_1 \cdot \mathbf{H}_1 + \mathbf{E}_2 \cdot \mathbf{H}_2\right) \\ - (b_R - c_R)\left(\mathbf{E}_1 \cdot \mathbf{H}_2 - \mathbf{E}_2 \cdot \mathbf{H}_1\right) \le 0, \end{aligned} \tag{15}$$

for every $\omega > 0$, where the subscripts R and I denote the real and the imaginary part. The arbitrariness of $\mathbf{E}_1, \mathbf{E}_2, \mathbf{H}_1, \mathbf{H}_2$ implies that

$$a_I \ge 0, \qquad d_I \ge 0, \tag{16}$$

$$(b_I + c_I)^2 + (b_R - c_R)^2 \le 4 a_I\, d_I. \tag{17}$$

These conditions are also sufficient for (15) to hold.

Apply now these conditions to (14). Inequalites (16) become

$$\begin{aligned} 0 \le a_I &= \frac{\epsilon_I\left[1 - \omega^2\left(\alpha_R\beta_R - \alpha_I\beta_I\right)\right] + \epsilon_R\omega^2\left(\alpha_I\beta_R + \alpha_R\beta_I\right)}{\left|1-\omega^2\alpha\beta\right|^2}, \\ 0 \le d_I &= \frac{\mu_I\left[1 - \omega^2\left(\alpha_R\beta_R - \alpha_I\beta_I\right)\right] + \mu_R\omega^2\left(\alpha_I\beta_R + \alpha_R\beta_I\right)}{\left|1-\omega^2\alpha\beta\right|^2}. \end{aligned}$$

If, further, the chiral coefficients α, β are real, then (16) and (17) become

$$\begin{gathered} \frac{\epsilon_I}{1-\omega^2\alpha\beta} \ge 0, \qquad \frac{\mu_I}{1-\omega^2\alpha\beta} \ge 0, \\ \omega^2\left[(\alpha\mu_R - \beta\epsilon_R)^2 + (\alpha\mu_I + \beta\epsilon_I)^2\right] \le 4\epsilon_I\,\mu_I. \end{gathered}$$

For small values of the chiral coefficients, namely when $\omega^2\alpha\beta < 1$, we get $\epsilon_I \ge 0$ and $\mu_I \ge 0$ as for dissipative dielectric and magnetic solids. If the bodies are nondissipative ($\epsilon_I = 0, \mu_I = 0$), then we have

$$\alpha\mu = \beta\epsilon.$$

This shows that the DBF model (1), where $\alpha = \epsilon\lambda$ and $\beta = \mu\lambda$, is compatible with thermodynamics.

5. MATERIALS WITH HIGHER-ORDER EFFECTS

Motivated by the model of isotropic and nongyrotropic media (cf. [7]), we look for constitutive equations of the form

$$\mathcal{D} = \epsilon\mathcal{E} + \alpha\nabla\times\mathcal{E} + \nabla\times(\nu\nabla\times\mathcal{E}) + \nabla(\eta\nabla\cdot\mathcal{E}), \tag{18}$$

$$\mathcal{B} = \mu\mathcal{H} + \beta\nabla\times\mathcal{H} + \nabla\times(\xi\nabla\times\mathcal{H}) + \nabla(\zeta\nabla\cdot\mathcal{H}). \tag{19}$$

In the time domain, these equations may be written, e.g., as

$$\mathbf{D} = \tilde{\epsilon}*\mathbf{E} + \tilde{\alpha}*\nabla\times\mathcal{E} + \nabla(\times\tilde{\nu}*\nabla\times\mathcal{E}) + \nabla(\tilde{\eta}*\nabla\cdot\mathcal{E}).$$

Also, $\epsilon^0, \epsilon'(\cdot), \dots,$ and hence $\epsilon, \dots$ are allowed to depend on the position so that the body is allowed to be inhomogeneous.

The degree of arbitrariness allowed by Maxwell's equations may be established as follows. Let $\eta, \zeta \neq 0$. At any point, the values of $\mathcal{E}, \mathcal{H}$ and of $\mathcal{D}, \mathcal{B}$, and hence, of $\nabla\times\mathcal{H}$, $\nabla\times\mathcal{E}$ by (9), can be chosen arbitrarily. Correspondingly, the values of $\nabla(\nabla\cdot\mathcal{E})$ and $\nabla(\nabla\cdot\mathcal{H})$ are given by (18) and (19). Also, the constraint on $\nabla\cdot\mathcal{B}$ and $\nabla\cdot\mathcal{D}$ is satisfied through appropriate values of $\Delta(\nabla\cdot\mathcal{E})$ and $\Delta(\nabla\cdot\mathcal{H})$.

Inequality (5) can be written as

$$\begin{aligned}&\Re\{i\omega\,[\epsilon\mathcal{E}^*\cdot\mathcal{E} + \alpha\mathcal{E}^*\cdot(\nabla\times\mathcal{E}) + \mathcal{E}^*\cdot\nabla\times(\nu\nabla\times\mathcal{E}) + \mathcal{E}^*\cdot\nabla(\eta\nabla\cdot\mathcal{E})\mu\mathcal{H}^*\\ &\cdot\mathcal{H} + \beta\mathcal{H}^*\cdot(\nabla\times\mathcal{H}) + \mathcal{H}^*\cdot\nabla\times(\xi\nabla\times\mathcal{H}) + \mathcal{H}^*\cdot\nabla(\zeta\nabla\cdot\mathcal{H})] - 2\nabla\cdot\mathcal{N}\} \leq 0,\end{aligned} \tag{20}$$

where $\mathcal{N}$ is such that $\Re\mathcal{N} = \overline{\mathbf{N}}$. Inequality (20) holds for arbitrary values of $\mathcal{E}, \nabla\times\mathcal{E}$, $\mathcal{H}, \nabla\times\mathcal{H}$ only if $\alpha = 0$ and $\beta = 0$.

Observe that

$$\begin{aligned}\mathcal{E}^*\cdot[\nabla\times(\nu\nabla\times\mathcal{E}) + \nabla(\eta\nabla\cdot\mathcal{E})] = \nabla\cdot[\nu(\nabla\times\mathcal{E})\times\mathcal{E}^*] + \nu(\nabla\times\mathcal{E})\cdot(\nabla\times\mathcal{E}^*)\\ + \nabla\cdot[\eta(\nabla\cdot\mathcal{E})\mathcal{E}^*] - \eta(\nabla\cdot\mathcal{E})\cdot(\nabla\cdot\mathcal{E}^*)\end{aligned}$$

and the like for $\nabla\times(\xi\nabla\times\mathcal{H}) + \nabla(\zeta\nabla\cdot\mathcal{H})$. Accordingly, inequality (20) can be given the form

$$\begin{aligned}&\Re\{i\omega[\epsilon\mathcal{E}^*\cdot\mathcal{E} + \nu(\nabla\times\mathcal{E})\cdot(\nabla\times\mathcal{E}^*) - \eta(\nabla\cdot\mathcal{E})(\nabla\cdot\mathcal{E}^*)\\ &\mu\mathcal{H}^*\cdot\mathcal{H} + \xi(\nabla\times\mathcal{H}^*)\cdot(\nabla\times\mathcal{H}) - \zeta(\nabla\cdot\mathcal{H}^*)(\nabla\cdot\mathcal{H})\\ &+\nabla\cdot(\nu(\nabla\times\mathcal{E})\times\mathcal{E}^* + \eta(\nabla\cdot\mathcal{E})\mathcal{E}^* + \xi(\nabla\times\mathcal{H})\times\mathcal{H}^* + \zeta(\nabla\cdot\mathcal{H})\mathcal{H}^*)] - 2\nabla\cdot\mathcal{N}\} \leq 0.\end{aligned} \tag{21}$$

Hence, it follows that

$$\epsilon_I \geq 0, \quad \mu_I \geq 0, \quad \nu_I \geq 0, \quad \xi_I \geq 0, \quad \eta_I \leq 0, \quad \zeta_I \leq 0$$

as $\omega > 0$ and

$$\mathcal{N} = \frac{1}{2}[\nu(\nabla\times\mathcal{E})\times\mathcal{E}^* + \eta(\nabla\cdot\mathcal{E})\mathcal{E}^* + \xi(\nabla\times\mathcal{H})\times\mathcal{H}^* + \zeta(\nabla\cdot\mathcal{H})\mathcal{H}^*].$$

These conditions, along with the vanishing of α and β are also sufficient for the compatibility of (18) and (19) with the second law in the form (5).

6. CONCLUSIONS

Models for optical activity, which account also for (dissipative) memory effects, are considered and compatibility with thermodynamics is investigated. As particular cases, the DBF model turns out to be compatible with thermodynamics while the model (18),(19) holds only if $\alpha = 0$,

$\beta = 0$. This is due to the fact that, by (18),(19), $\nabla \times \mathcal{E}$ and $\nabla \times \mathcal{H}$ may take arbitrary values. Now by (13), subject to (14), in the simpler case $\alpha\mu = \beta\epsilon =: \psi$, we have

$$\mathcal{D} = \frac{\epsilon}{\delta}\mathcal{E} + i\omega\frac{\psi}{\delta}\mathcal{H}, \qquad \mathcal{B} = -i\omega\frac{\psi}{\delta}\mathcal{E} + \frac{\mu}{\delta}\mathcal{H}. \tag{22}$$

Consequently, if ψ/δ is real, then (17) holds identically and any value of ψ is allowed. This shows that compatibility with thermodynamics is allowed if $\mathcal{H}$ affects $\mathcal{D}$ and $\mathcal{E}$ affects $\mathcal{B}$ as in (22). It is of interest that such is the case for the DBF model as well as for the Condon model [8]

$$\mathbf{D} = \epsilon\mathbf{E} - \kappa\dot{\mathbf{H}}, \qquad \mathbf{B} = \mu\mathbf{H} + \kappa\dot{\mathbf{E}},$$

applied to time-harmonic fields, and the Chambers model [9]

$$\mathbf{D} = \epsilon\mathbf{E} + \chi\mathbf{H}, \qquad \mathbf{B} = \mu\mathbf{H} - \chi\mathbf{E}$$

if κ is real and χ is imaginary.

REFERENCES

1. A. Lakhtakia, *Beltrami Fields in Chiral Media*, World Scientific, Singapore, (1994).
2. G.A. Maugin, *Continuum Mechanics of Electromagnetic Solids*, North Holland, Amsterdam, (1988).
3. D. Jou, J. Casas-Vazquez and G. Lebon, *Extended Irreversible Thermodynamics*, Springer, Berlin, (1996).
4. B.D. Coleman and E.H. Dill, Thermodynamic restrictions on the constitutive equations of electromagnetic theory, *ZAMP* **22**, 691–702, (1971).
5. M. Fabrizio and A. Morro, Thermodynamics of electromagnetic isothermal systems with memory, *J. Non-Equilib. Thermodyn.* **10**, 110–128, (1997).
6. M. Fabrizio and A. Morro, Dissipativity and irreversibility of electromagnetic systems, *Math. Mod. Meth. Appl. Sci.* **10**, 217–246, (2000).
7. V.M. Agranovich and V.L. Ginzburg, *Crystal Optics with Spatial Dispersion, and Excitons*, Springer, Berlin, (1984).
8. E.U. Condon, Theories of optical rotatory power, *Rev. Mod. Phys.* **9**, 432–457, (1937).
9. L.G. Chambers, Propagation in a gyrational medium, *Quart. J. Mech. Appl. Math.* **9**, 360–370, (1956).